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Author manuscript

J Environ Qual. Author manuscript; available in PMC 2016 April 27.

Published in final edited form as:

J Environ Qual. 2014 November; 43(6): 1873–1885. doi:10.2134/jeq2014.05.0227.

## Meteorological Influences on Nitrogen Dynamics of a Coastal Onsite Wastewater Treatment System

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#### **Abstract**

Onsite wastewater treatment systems (OWTS) can contribute nitrogen (N) to coastal waters. In coastal areas with shallow groundwater, OWTS are likely affected by meteorological events. However, the meteorological influences on temporal variability of N exports from OWTS are not well documented. Hydrogeological characterization and seasonal monitoring of wastewater and groundwater quality were conducted at a residence adjacent to the Pamlico River Estuary, North Carolina during a two-year field study (October 2009-2011). Rainfall was elevated during the first study year, relative to the annual mean. In the second year, drought was followed by extreme precipitation from Hurricane Irene. Recent meteorological conditions influenced N speciation and concentrations in groundwater. Groundwater total dissolved nitrogen (TDN) beneath the OWTS drainfield was dominated by nitrate during the drought; during wetter periods ammonium and organic N were common. Effective precipitation (P-ET) affected OWTS TDN exports because of its influence on groundwater recharge and discharge. Groundwater nitrate-N concentrations beneath the drainfield were typically higher than 10 mg/l when total bi-weekly precipitation was less than evapotranspiration (precipitation deficit: P<ET). Overall, groundwater TDN concentrations were elevated above background concentrations at distances >15 m downgradient of the drainfield. Although OWTS nitrate inputs caused elevated groundwater nitrate concentrations between the drainfield and the estuary, the majority of nitrate was attenuated via denitrification between the OWTS and 48 m to the estuary. However, DON originating from the OWTS was mobile and contributed to elevated TDN concentrations along the groundwater flowpath to the estuary.

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## Introduction

Excess nutrient inputs have resulted in impairments to the majority of US coastal rivers and bays (Howarth et al. 2002). In coastal settings, nitrogen (N) is of particular concern because eutrophication in most estuaries in the temperate zone is N-limited (Howarth and Marino 2006) and increases in N inputs have led to increased harmful algal blooms and eutrophic conditions (Rabalais et al. 2009). Major N inputs are from point and non-point sources, typically agricultural fertilizer, animal manure, atmospheric deposition, and effluents from wastewater treatment systems (Howarth et al. 2002). Although nutrient management efforts have focused primarily on agricultural inputs (Heathwaite et al. 2000), in many coastal areas research findings have shown that wastewater may be the dominant N source that can affect groundwater and surface water quality (Valiela et al. 1997; Kroeger et al. 2006a). Estimates of N inputs to coastal watersheds in the eastern U.S. suggest that residential on-site wastewater treatment systems (OWTS, also known as septic systems) may contribute from 4–64 % of the N loading (McClelland and Valiela, 1998; Pradhan et al. 2007). Watershed-scale estimates of N inputs from OWTS have been challenging to obtain because N removal from conventional OWTS can be highly variable across space and time.

Although much work has been focused on OWTS N exports, it is not well understood how rainfall and groundwater levels affect OWTS N treatment. Nutrient leaching and transport is generally enhanced in settings immediately adjacent to water bodies, with sandy soils, and elevated water tables (Carey et al. 2013). Our objective was to quantify N transport from an OWTS in a sandy coastal surficial aquifer adjacent to a nutrient-sensitive estuary and evaluate the meteorological controls on N exports. The goals were to assess how weather-related effects on water table depth and separation distance influence OWTS N treatment and to quantify the effects of subsurface wastewater inputs on nitrogen fate and transport along a groundwater flowpath to an estuary.

In a review of eight published studies, Valiela et al. (1997) reported that OWTS can retain from 10–90% of N. In situations when N-retention is low, exports from OWTS can affect human and ecological health, locally by affecting drinking water supplies (Harman et al 1996) and regionally by contributing to nutrient enrichment of surface waters (Valiela et al. 1997). In coastal settings where sandy soils predominate and water tables are shallow (Fan et al. 2013), conventional OWTS are likely to contribute to elevated nitrate (NO<sub>3</sub><sup>-</sup>) concentrations to the shallow groundwater and surface water (Gold and Sims 2001). This is significant because water quality of coastal estuaries can be affected by N inputs at concentrations lower than the National Primary Drinking Water Maximum Contaminant Levels of 10 mg/l NO<sub>3</sub><sup>-</sup>-N (Environmental Protection Agency (USEPA) (2009), and N concentrations at levels above 2 mg/L as NO<sub>3</sub><sup>-</sup>-N may contribute to eutrophic conditions and may be toxic to certain aquatic organisms (USEPA 2000; Gold and Sims 2001; Camargo et al. 2005).

OWTS consist of a septic tank, drainfield, and the native soils. Organic and ammonium  $(NH_4^+)$  forms of N are commonly the dominant forms in the raw wastewater entering the septic tank. The anaerobic environment of the septic tank favors ammonification and ammonium becomes the dominant N-form in its effluent. Conventional OWTS tanks are not

generally designed for treating N, thus they remove only a small fraction of the N (10–20%) from wastewater before disposal to the subsurface (Oakley et al. 2010). The majority of N treatment typically occurs in the soils as the effluent percolates through the unsaturated zone and most of the N is expected to convert to the nitrate form. In subsurface anaerobic environments that contain denitrifying bacteria and organic matter, denitrification can lower NO<sub>3</sub>-concentrations in soil water and groundwater (Wilhelm et al. 1994a). Since OWTS wastewater is often discharged at depths of ~1 m in the subsurface, plant uptake and denitrification in the shallow organic soils is generally minimal (Gold and Sims 2001). Studies conducted on systems located in sandy soils show that OWTS NO<sub>3</sub><sup>-</sup> may cause elevated NO<sub>3</sub><sup>-</sup> concentrations in groundwater (Robertson et al., 1991; Humphrey et al. 2010a). The extent of NO<sub>3</sub><sup>-</sup> contamination is variable and is a function of a number of controls including: water table depth, soil texture, OWTS density, distance from the drainfield, groundwater flow velocity, dilution/dispersion, biomat formation, climate and seasonal factors, presence of riparian buffers, age and design of the system, water use, household practices, and system maintenance (Beal et al. 2005, Gold and Sims 2001, Valiela et al. 1997, Oakley et al. 2010).

Although numerous studies have focused on the effects of soil type and groundwater depth on OWTS N treatment (Cogger and Carlile 1984; Karathanasis et al. 2006; Humphrey et al. 2010a) few studies have focused on the effects of meteorological conditions on OWTS N treatment in coastal settings. Arnade (1999) looked at the effects of increased summer rainfall patterns and shallow water tables on groundwater NO<sub>3</sub><sup>-</sup>-N concentrations in Palm Bay, FL. She found that NO<sub>3</sub><sup>-</sup>-N concentrations in groundwater generally decreased with distance from the drainfield, but the decline was less pronounced during the wet season. For 15 systems in the North Carolina Coastal Plain, Cogger and Carlile (1984) found that groundwater N species and concentrations were influenced by seasonal variations of the water table. The systems that were continually saturated had the poorest performance, and the greatest transport of N occurred for systems located in areas with high gradients and continuous soil saturation. Those systems tended to export more NH<sub>4</sub><sup>+</sup> but less NO<sub>3</sub><sup>-</sup>, relative to better drained systems. In a one-year study, LaPointe et al. (1990) performed monthly sampling of N in surface water and groundwater in a limestone aquifer in the Florida Keys and found elevated N linked to OWTS. They found elevated N concentrations in surface waters during summer months presumably due to increased groundwater nutrient inputs, since hydraulic gradients can be increased during the summer wet season. Although rainfall did not show a strong correlation with groundwater nutrient concentrations, potential evapotranspiration was significantly correlated with groundwater nutrient concentrations, presumably due to the influence of evaporative losses on groundwater recharge potential. Overall, these studies indicated that weather patterns can affect OWTS N treatment and the fate and transport of N in surficial aquifers.

In coastal areas, climate change and the resultant sea level rise has the potential to affect OWTS N treatment. There is growing concern that changes in temperature and precipitation patterns (USEPA 2012a) and sea level rise (Werner and Simmons 2009; USEPA 2012b) may affect the efficiency of conventional OWTS to remove/retain N prior to discharge to groundwater and eventually surface waters. A better understanding of OWTS N contributions to coastal aquifers is needed to improve model estimates, evaluate potential

impacts to nutrient sensitive estuaries, and to help design and implement OWTS that minimize the risks to human and ecosystem health.

## **Materials and Methods**

#### Site Selection

We selected a year-round residential site adjacent to the nutrient sensitive waters of the Pamlico River Estuary in coastal Beaufort County, North Carolina in 2009. We selected this site because it was directly upgradient of the estuary, had a sandy surficial aquifer that was connected to the estuary, and based on initial study (Deal et al. 2011) it was compliant with the North Carolina setback and separation distance regulations (NC DHHS 1990) (Figure 1.a.). In addition, the homeowners were willing to allow soil coring and groundwater monitoring at their residence.

## Site characterization and orientation of subsurface wastewater plume

During initial site characterization studies (Humphrey et al. 2010b) the OWTS components, including the septic tank and drainfield trenches, were located by use of a tile drain probe and the orientation of the subsurface wastewater plume was estimated by use of an OhmMapper TR1 electrical resistivity mapper (Geometrics, Inc. San Jose, CA). Resistivity declines adjacent to subsurface wastewater disposal sites have been used to infer the location of wastewater-affected groundwater in Mountain (Roy et al. 2008) and Coastal Plain settings (Smith 2013). During the current study, we generated a two-dimensional electrical resistivity profile from an electrical resistivity survey conducted in February 2011 to confirm the influence of wastewater on groundwater quality downgradient of the drainfield. The OhmMapper transmitter and receiver were separated by 2.5, 5, and 10 m and towed over the transect line 3 times using 5 m dipoles. By spacing the transmitter and receiver at greater separation spacings, the current penetration depth increased, which allowed to image to depths below the water table (deeper than 1.5 m). The surveys provided apparent resistivity data for the subsurface along the transect line. We inverse modeled the apparent resistivity data with RES2DINV software<sup>TM</sup> (Geotomo Software, 2014). We compared the earth resistivity data with groundwater specific conductivity data collected in piezometers on February 21, 2011 (Figure 1.b.). The decline in resistivity and increase in groundwater conductivity adjacent to, and downgradient from, the drainfield suggested the approximate location of wastewater-affected groundwater. However, it should be noted that directly adjacent to the estuary, salinity may also result in increased groundwater conductivity.

We estimated the direction of groundwater flow at the site to be in the southerly direction towards the estuary, from a three-point problem solution at the residence (Humphrey et al. 2010b; Humphrey et al. 2013). The tile drain probe survey revealed that the OWTS drainlines were buried at approximately 60 cm depth below the land surface. We installed piezometers and lysimeters upgradient of the drainfield, in the drainfield, and at various distances downgradient from the drainfield on and adjacent to the projected groundwater flowpath to the estuary. We installed a total of 28 piezometers up- and down-gradient of the OWTS groundwater flowpath for groundwater sample collection and monitoring (Figure 1.a.) at depths of 1.3 to 3.7 m. During the second year of study (2011) we installed six

porous cup tension lysimeters as follows: one group at 30, 45, and 60 cm depth in the drainfield and another group at the same depths in an area upgradient of the drainfield. We used the lysimeters to collect soil water quality data at bi-monthly intervals in the drainfield and upgradient soils.

#### Determination of water table depth

We determined bi-monthly depth to water-table measurements with a Solinst Model 107 Temperature Level and Conductivity (TLC) meter (Solinst Canada Ltd., Georgetown, ON). On each sampling date, we estimated the separation distance between the water table and the drainlines as the difference between the groundwater depth at piezometer 4s and the depth to the drainlines. In piezometers 3 and 5, near the drainfield disposal trenches, we installed automated Hobo U-20 water level loggers (Onset Computer Corp., Bourne, MA) programmed to record groundwater levels every 0.5 hours. We used the automated water level measurements to observe temporal vertical separation distance (trench bottom and water table) and water table dynamics. In addition, we installed groundwater level loggers in piezometers 2, 3, 13, and 15, away from the drainfield (Figure 1.a.). To determine groundwater and septic tank pH, specific conductivity, temperature, and dissolved oxygen concentrations in groundwater and wastewater at the site we used a calibrated YSI 556 field meter (YSI Inc., Yellow Springs, OH). In addition, at piezometers 5 (adjacent to the drainfield) and 2 (upgradient from the drainfield), we used two YSI 6920 data logging sondes to record groundwater specific conductivity at a 30-minute interval. We obtained rainfall data from a weather station (Warren Field- 17 km to the northwest in Washington, NC) through the NC CRONOS database for the period of 2009–2011. Monthly reference crop evapotranspiration (ET) was estimated using the Penman-Monteith method (Monteith, 1965) at the Tidewater Research Station, Plymouth, NC (approximately 40 km to the north) (State Climate Office of North Carolina, 2014a). Typically, ET in the region is elevated during the crop growing season (April-October) and minimal during the dormant season (November-March) (USDA 1997, Sun et al. 2002).

#### Sampling and analyses of wastewater and groundwater samples

We sampled the septic tank monthly from October 2009 to May 2010 and from January 2011 to October 2011. We collected groundwater samples from piezometers and surface water samples from the estuary bi-monthly from November 2009 to May 2010 and from January to October 2011 (Figure 1.a.). A gap in funding caused the sampling pause between May 2010 and January 2011. We used a new bailer for collecting groundwater samples from each piezometer and purged each piezometer prior to sampling. We kept samples on ice and delivered them to the East Carolina University Central Environmental Laboratory (CEL) within 12 hours where laboratory staff filtered the samples prior to nitrogen analyses. Ammonia was analyzed by use of the Solorzano method (Standards Methods for the Examination of Water and Wastewater, 1995). Dissolved Kjeldahl N, nitrate/nitrite, and chloride were analyzed by use of a SmartChem 200 discrete wet chemistry analyzer (Westco Scientific Instruments Incorporated, Brookfield, Connecticut).

To estimate the effect of dilution on the reduction of groundwater total dissolved N (TDN) concentrations in the drainfield, we used a two-component mixing model using the end

members of wastewater and background groundwater TDN and chloride (Cl<sup>-</sup>) concentrations, based on the following equation:

$$Cgwm = Cww * (1 - fgw) + Cgwb * fgw$$
 (Eq. 1)

where:

Cgwm is the measured groundwater concentration (mg/l)

Cww is the wastewater concentration (mg/l)

fgw is the fraction of groundwater, and

Cgwb is the background groundwater concentration (mg/l)

We collected groundwater samples for isotopic analysis from piezometers on four dates: 1/25/10; 5/24/10; 2/18/11; and 6/6/11. The University of California, Davis Stable Isotope Facility conducted the isotopic analysis (UC Davis, 2013). They analyzed groundwater samples for  $^{15}N$  and  $^{18}O$  in  $NO_3^-$  using a ThermoFinnigan Gas Bench plus PreCon trace gas concentrations system interfaced to a ThermoScientific Delta V Plus isotope-ratio mass spectrometer (Bremen, Germany). The Stable Isotope Facility's staff used the methods of Sigman et al. (2001), Casciotti et al. (2002), and Granger and Sigman (2009), for the analyses of  $^{15}N$  and  $^{18}O$  in  $NO_3^-$ .

## Statistical analysis

We used non-parametric Mann-Whitney tests (Conover and Iman1981; Conover 1999) to compare the distributions of groundwater TDN concentration for UG (upgradient) groundwater vs. groundwater affected by the OWTS (DF, GW<15m, GW>15m). We adjusted the significance threshold to address potential Type I errors using the Bonferroni method recommended by Abdi (2007) and conducted all statistical analyses using Minitab v. 16 (Minitab Incorporated, State College, Pennsylvania). The Bonferroni method is one of the methods used to adjust multiple comparisons. We used the 15 m distance because it corresponds to the North Carolina minimum surface water setbacks for OWTS (NC DHHS, 1990).

## **Results and Discussion**

#### **OWTS Effects on Groundwater Nitrogen Concentrations**

Electrical resistivity patterns and groundwater specific conductivity patterns suggested that wastewater inputs caused elevated total dissolved solids in the surficial aquifer and that a wastewater plume extended from the drainfield to the estuary (Figure 1.b.). The mean groundwater specific conductivity and TDN values for the duration of the study were elevated along the projected groundwater flowpath to the estuary (Table 1.a.). The septic tank had elevated specific conductivity, temperature, and TDN relative to groundwater at the site. In the tank, DON was the dominant species, NH<sub>4</sub><sup>+</sup>-N was also elevated, but NO<sub>3</sub><sup>-</sup>-N was generally absent. Groundwater upgradient (UG) of the OWTS drainfield had low concentrations of TDN (generally present as NH<sub>4</sub><sup>+</sup> and DON), low specific conductivity, and elevated dissolved oxygen concentrations, compared to groundwater adjacent to and

downgradient from the drainfield. As groundwater flowed below the OWTS drainlines, dissolved oxygen declined, specific conductivity increased, and all N species increased. The dominant N-species in the drainfield (DF) groundwater was  $NO_3^-$ . At greater distances from the drainfield, the N-species concentrations declined. Table 1.b. shows results of non-parametric Mann-Whitney tests (Conover and Iman 1981, Conover 1999) adjusted for multiple comparisons using the Bonferroni method (Abdi, 2007). Tests revealed that  $NO_3^-$ -N, DON, and TDN concentration distributions were significantly greater (p<0.0125) in the DF groundwater and GW<15 m from the drainfield than the UG groundwater. As distance from the drainfield increased, the differences between UG groundwater concentrations and groundwater concentrations downgradient of the OWTS drainfield decreased. However, Mann-Whitney tests adjusted for multiple comparisons indicated that DON and TDN concentration distributions were still significantly greater (p<0.0125) in the DF, GW<15 m, and GW>15 m than the UG groundwater (Table 1.b.).

#### Influences on Drainfield Groundwater Total Dissolved Nitrogen Concentrations

Drainfield groundwater TDN concentrations were influenced by the loading of N from the tank, the vertical separation distance between the water table and the drainlines (Figure 2.a.), and the difference between precipitation and evapotranspiration (P-ET) (Figure 2.b.). There was a strong inverse correlation between the amount of precipitation excess and drainfield groundwater TDN concentration (Pearson correlation coefficient=-0.73; p=0.025). During periods of precipitation deficit (precipitation</a>evapotranspiration), median drainfield groundwater TDN concentrations were generally elevated (33.8 mg/l). In contrast, during periods of precipitation excess (precipitation>evapotranspiration), median drainfield groundwater TDN concentrations were generally lower (9.7 mg/l) and the distributions were significantly different (p=0.046) than during periods of precipitation deficit (Figure 2.c.).

The vertical separation distance between the water table and the drainlines was influenced by recent meteorological conditions (Figure 3.a.). When precipitation excess occurred, the separation distance was smaller. During extreme wet conditions (following Hurricane Irene) the groundwater was elevated above the drainlines following a flood event that resulted in a negative separation distance. In contrast, during the drought conditions that occurred in the spring and summer of 2011, the separation distances increased up to approximately 1 m. In North Carolina, the required separation distance for group I sandy soils is 45 cm, and this requirement was met for 4 of 9 sampling dates. When the conditions were wetter (precipitation excess) the separation distance was more likely to be less than the NC required 45 cm. Separation distance had an influence on N speciation in drainfield groundwater; the dominant N species was NO<sub>3</sub><sup>-</sup> during the second year of study when drought conditions were common. NH<sub>4</sub><sup>+</sup> and DON made up a larger percentage of the TDN during the wetter first year. We compared these data to those from a previous study of 16 OWTS in coastal North Carolina (Humphrey et al. 2010a) and found a similar pattern of increasing NO<sub>3</sub><sup>-</sup> dominance with increased separation distance (Figure 3.b.).

The relationship between greater separation distance and increased nitrification has been documented in the conceptual model of Wilhelm et al. (1994b) and in several field studies in coastal North Carolina (Cogger et al. 1988, Humphrey et al. 2010a). In their study of the

effects of water table variability on OWTS N treatment, Cogger et al. (1988) found that NH<sub>4</sub><sup>+</sup> was dominant at locations with shallow water tables, and NO<sub>3</sub><sup>-</sup> dominated at locations where the water table was deeper and separation distance was greater than 30 cm. At shallow groundwater locations, during dry periods the presence of NO<sub>3</sub><sup>-</sup> increased suggesting that the seasonal variations in aerobic conditions could lead to greater NO<sub>3</sub><sup>-</sup>-N concentrations in groundwater during drier conditions, presumably due to increased nitrification. Although these studies did not directly show the link between precipitation deficits and nitrification, an earlier study by Starr and Sawhney (1980) monitored the effects of a septic system on drainfield soil water N concentrations over a two year period. They found that during a year with greater rainfall, NH<sub>4</sub><sup>+</sup> was more mobile and little nitrification occurred. During the following year, 50% less rainfall occurred and the drier conditions led to increased nitrification and a pulse of NO<sub>3</sub><sup>-</sup> in the drainfield. They suggested that rainfall amounts can have an indirect control on N mobility, due to the influence on N-speciation.

## **Nitrogen Speciation and DON Mobility**

In the septic tank, wastewater TDN was dominated by DON, with lesser concentrations of NH<sub>4</sub><sup>+</sup>-N. In the soil water below the drainfield, NO<sub>3</sub><sup>-</sup>-N was the dominant N species and DON and NH<sub>4</sub><sup>+</sup>-N were present at low concentrations providing evidence of effective nitrification in the drainfield soils (Table 1.a.). However, during wet periods that occurred in the first year of study, NH<sub>4</sub><sup>+</sup>-N and DON were elevated in the groundwater beneath the drainfield, relative to NO<sub>3</sub><sup>-</sup>-N (Figure 4). Along the groundwater flowpath from the drainfield to the estuary, NO<sub>3</sub><sup>-</sup>-N was present at the highest concentrations, particularly during drought conditions in year 2 (Figure 4). At distances greater than 15 m downgradient from the drainfield Mann-Whitney tests adjusted by the Bonferroni method for multiple comparisons revealed that only DON and TDN concentrations were significantly greater (p<0.0125) than background concentrations (Table 1.b.).

Our data suggested that groundwater NH<sub>4</sub><sup>+</sup>-N concentrations would decline to background concentrations prior to discharge to the estuary, but DON and to a lesser extent NO<sub>3</sub><sup>-</sup>-N could still be elevated relative to background concentrations. Interestingly, the groundwater DON concentrations were elevated during the first (wet) year of study (median concentration=1.33 mg/l) and lower during the second (drought) year of study (median concentration = 0.7 mg/l). This suggests that during wetter periods organic matter was not breaking down as efficiently (Wilhelm et al. 1994b). Although the groundwater DON data were somewhat variable and did show a large decline in the first 20 m downgradient from the drainfield, DON concentrations greater than 20 m downgradient were still elevated above background groundwater DON concentrations, suggesting a wastewater source (Figure 4). This additional dissolved nitrogen contributes to elevated TDN at the site. The DON data suggest that there is a component of DON from the septic tank that is relatively mobile in groundwater. DON was typically the primary form of dissolved N in wastewater in the tank at this study site (mean DON= 68 mg/l; mean TDN= 86 mg/l). In their review, McCray et al. (2005) documented median DON values of 14 mg/l (n=6) and NH<sub>4</sub><sup>+</sup>-N values of 58 mg/l (n=37) for tank effluent from a number of studies. In the current study, the tank was pumped prior to monitoring, it is possible that the removal of sludge could affect the conversion of organic N to NH<sub>4</sub><sup>+</sup> in the tank we studied, relative to those studied by McCray et al. (2005).

Similarly to McCray et al. (2005), Withers et al. (2011) found a range of 0–13.9 mg/l DON in tank effluent. DON is expected to be converted to inorganic forms and assumed to be a minor constituent in groundwater adjacent to OWTS (McCray et al. 2005 and Reay 2004).

Several recent studies suggest that TDN and DON in wastewater effluent in coastal North Carolina may be elevated compared to other regions. In a study that sampled 10 septic tanks for TDN in eastern North Carolina (Carteret County), Humphrey (2009) also found elevated DON in tanks, on average wastewater had more than 80 mg/l of DON. In addition, a study by Berkowitz (2007), suggested that wastewater effluent in coastal North Carolina tends to have elevated TDN compared to many other locations throughout the nation. His study showed median Total Kjeldahl N values for effluent of approximately 80 mg/l. In a recent study DON effluent was evaluated at 4 biological nutrient removal wastewater treatment plants, including one on the Neuse River, North Carolina (Sattayatewa et al. 2010). They found that treatment plants could remove 60–80% of DON, but effluent contained between 0.5–2 mg/l DON. This range of effluent DON is similar to the range of groundwater DON we observed.

Elevated DON concentrations in surface waters have recently gained interest, as they may play an important role in supplying N nutrition to phytoplankton and bacteria (Berman and Bronk 2003). In Cape Cod, Kroeger et al. (2006b) suggested that DON could be the dominant form of N transport in coastal watersheds, and in some watersheds DON was contributed by anthropogenic sources (i.e. wastewater). Recent studies suggest that DON from anthropogenic sources may be more bioavailable to microbes and aquatic bacteria than DON from natural sources (various sources as cited in Pellerin et al. 2006). If DON compounds are biologically available they can affect the population dynamics and species diversity in aquatic ecosystems. Future work is needed to characterize the compounds that make up the DON pool in wastewater and groundwater and evaluate how mobile and bioavailable these compounds are in groundwater and surface water bodies. This is an important issue in the nutrient-sensitive Tar-Pamlico and Neuse watersheds in North Carolina, where DON concentrations in these rivers have been trending upwards over the last two decades (Lebo et al. 2012) and nutrient-sensitive management strategies are being undertaken.

#### Nitrogen Decline with Distance and Setback Regulations

TDN concentrations in groundwater downgradient from the drainfield revealed an exponential decay pattern with distance from the drainfield. The general trends were consistent with those published by Valiela et al. 1997 (Figure 5). Generally, TDN concentrations in the drainfield and downgradient groundwater were higher during the second year of study when drought conditions were prevalent. Although the groundwater TDN concentration data were variable, distance from the drainfield was a significant predictor of TDN concentration (Figure 5). Historically, North Carolina has been prone to drought and drought conditions have occurred in the state during 5 of the last 10 years (State Climate Office of North Carolina 2014b). Considering that the EPA's maximum contaminant level for NO<sub>3</sub><sup>-</sup>-N in potable groundwater is 10 mg/l, these data suggest that during drought conditions this standard is likely to be exceeded in this sandy coastal aquifer.

Based on the calculated mixing line for Cl<sup>-</sup> and TDN, the groundwater Cl<sup>-</sup> concentrations measured beneath the drainfield suggested that dilution of wastewater by upgradient groundwater flowing underneath the drainfield could account for up to 60% of concentration declines (Figure 6.a.). During the drought conditions from April-August 2011, drainfield groundwater TDN concentrations were slightly elevated above the mixing line and proximity to the mixing line suggested that dilution played a role in TDN decline. On other dates, when TDN concentrations in drainfield groundwater fell below the mixing line, other mechanisms such as denitrification must have played a more important role. Drainfield groundwater TDN and Cl<sup>-</sup> concentration data were compared with the tank wastewater concentration data and these data suggested that dilution plays an important role in TDN decline. Median Cl<sup>-</sup> concentration declined by 35% from the tank to the drainfield and median TDN declined by 68%, suggesting other mechanisms are responsible for TDN concentration declines (Figure 6.b.). A similar TDN decline was evident in lysimeter data (Table 1.a.). At depths of 0.9–1.2 m, TDN concentrations declined from a median 94 mg/l in wastewater to a median of 26 mg/l (72% decline) in the soil water. The median Cl<sup>-</sup> decline between the wastewater and soil water for the same sampling dates was 38%. The wastewater, drainfield groundwater, and soil water data revealed that the majority of N retention/loss occurred in the shallow soils and biomat, prior to groundwater recharge. On the dates that groundwater was sampled (excluding the October 2011 date that was affected by Hurricane Irene flooding) median tank Cl<sup>-</sup> concentration was 93.0 mg/l and TDN concentration was 93.8 mg/l. A Mann Whitney test showed tank Cl<sup>-</sup> concentration and tank TDN concentration distributions were not significantly different (p>0.05); however the median groundwater Cl<sup>-</sup> in the drainfield was 60 mg/l and groundwater TDN was 29.4 mg/l and these were significantly different (p=0.03). These data suggest that approximately 33 mg/l of TDN concentration decline can be attributed to dilution and approximately 32 mg/l attributed to retention/loss of TDN between the tank and drainfield piezometers. The mechanisms responsible for this TDN concentration decline can include assimilation, cation exchange, and denitrification in the drainfield.

To further evaluate denitrification, groundwater dissolved oxygen and <sup>15</sup>N and <sup>18</sup>O in NO<sub>3</sub><sup>-</sup> patterns along the groundwater flowpath from the drainfield to the estuary were studied. The patterns suggest that denitrification plays a significant role in groundwater NO<sub>3</sub><sup>-</sup>-N declines (Figure 7). The relationship between <sup>15</sup>N and <sup>18</sup>O in NO<sub>3</sub><sup>-</sup> suggested a wastewater NO<sub>3</sub><sup>-</sup> source in the drainfield and isotopic enrichment along the flowpath from the drainfield to the estuary, particularly for piezometers 4, 5, 6, 7, 8, 10, and 16. These piezometers generally had elevated groundwater specific conductivity (Figure 1) and TDN concentrations, indicating the core of the wastewater plume. Based on the fractionation relationships provided by Chen et al. (2009) and Silva et al. (2002), the median <sup>15</sup>N and <sup>18</sup>O in NO<sub>3</sub>-groundwater composition data suggest that denitrification accounts for approximately 20% of the groundwater NO<sub>3</sub><sup>-</sup>-N concentration reduction at the drainfield, and up to approximately 85% of the groundwater NO<sub>3</sub><sup>-</sup> concentration reduction at piezometers 10 and 16 close to the estuary (Figure 7.a.). The enrichment of  $NO_3^{-15}N$  and  $^{18}O$  in groundwater corresponded with a decrease in NO<sub>3</sub><sup>-</sup>-N concentrations in groundwater along the flowpath from the drainfield to the estuary (Figure 7.b.) consistent with a denitrification mechanism. Overall, the data suggested that due to dilution and denitrification, most of the groundwater

nitrate was attenuated. Although the groundwater  $NO_3^-$ -N concentration declines from the drainfield piezometers (median  $NO_3^-$ -N = 8.3 mg/l) along the groundwater flowpath to piezometer 10 (median  $NO_3^-$ -N = 1.1 mg/l) and piezometer 16 (0.15 mg/l), directly adjacent to the estuary are large, the groundwater discharging to the estuary still had slightly elevated  $NO_3^-$  concentrations above median background concentrations (0.03 mg/l).

#### **Seasonality of OWTS Nitrogen Exports**

On an annual basis, the average TDN load to the drainfield soils from the tank was 11.3 kg as N. Since there were two residents, this equated to approximately 5.7 kg-TDN/person-yr, similar to the per capita estimates of 5–6 kg-TDN/person-yr by the EPA (USEPA 2012b). However, because of attenuation processes in the soils and surficial aquifer, the groundwater TDN exports (assuming a 20 m by 3 m cross-sectional area) from the drainfield were much lower (mean drainfield exports 0.85 kg-TDN/yr). Based on the relationship between P-ET and groundwater TDN concentrations (Figure 2.b.) and groundwater flow rates (GW flow  $(L/month) = 407.1(P-ET cm) + 2094; R^2=0.80)$  the study data suggest that seasonal variations in groundwater TDN concentrations and groundwater flow should result in seasonally variable N exports from the drainfield (Figure 8.a.-c.). To further evaluate the effects of seasonal variations of precipitation excess on groundwater N exports, P and ET data collected at the Tidewater Research Station, Plymouth NC from 2004-2012 were analyzed for seasonal patterns. These data revealed that precipitation deficits were common during the growing season (April-October; median P-ET = -3.77 cm) and precipitation excess was common during the dormant season (November-March; median P-ET=0.40 cm) (Figure 8.a.). Based on the relationships at the site between P-ET, drainfield groundwater TDN, and groundwater flow rates, the 2004–2012 P-ET patterns suggested that during the dormant season TDN concentrations should be lower but groundwater flow rates higher (Figure 8.b.). These relationships suggest an increase in TDN exports (Figure 8.c.) during the dormant season due to increased hydraulic head gradients and the resultant increase in groundwater discharge to the estuary.

The effects of recent weather patterns on groundwater levels can have an influence on both concentrations of TDN, due to the influence of dilution, and the mobility of TDN due to the effects of separation distance on N-speciation. The lack of groundwater recharge during periods of precipitation deficits caused the water table to decline, particularly during the growing season when ET rates are relatively high (approximately 70% of annual precipitation; Sun et al. 2002) in this region. The water table decline leads to increased separation distances, resulting in a thicker vadose zone and more aerated soils underneath the drainfield which promote nitrification. Generally, during drought periods, the highest NO<sub>3</sub><sup>-</sup>-N concentrations were observed in the drainfield because of increased nitrification and decreased dilution. Hantzche and Finnemore (1992) studied the effects of recharge on groundwater NO<sub>3</sub><sup>-</sup>-N concentrations adjacent to OWTS drainfields in three California communities (two in coastal settings). They developed a model that calculated the groundwater NO<sub>3</sub><sup>-</sup>-N concentrations as a function of wastewater loading and groundwater recharge and showed that increased levels of groundwater recharge can reduce the groundwater NO<sub>3</sub><sup>-</sup>-N concentrations adjacent to OWTS.

At coastal sites with year-round residents, if there is a seasonal reduction in groundwater recharge due to elevated growing season evapotranspiration (Sun et al. 2002), then a growing season increase in drainfield groundwater TDN concentrations may occur due to the decrease in recharge and decreased dilution. In our study, drainfield groundwater TDN concentrations were elevated during drought conditions and declined during the wetter periods. In addition, in coastal communities with a strong tourism base, there may be increased wastewater loading during the summer tourist season. For example, in a study of three seasonally used septic systems in coastal Rhode Island, Postma et al. (1992) found that for houses occupied from July to September there was a large increase in groundwater NO<sub>3</sub>--N concentrations (up to 115 mg/l) adjacent to septic systems and increases in concentration were observed at least 6 m from the systems. This work showed that in sandy coastal areas with shallow water tables, seasonal variability in groundwater N inputs can also be influenced by seasonal tourism.

#### **Conclusions**

Coastal surficial aquifers can transmit OWTS N that contributes to surface water N loading. During wetter periods, increased groundwater discharge can lead to greater nutrient loading to adjacent water bodies, even though groundwater TDN concentrations may be lower. Although nitrate is commonly considered the most mobile N species in groundwater affected by OWTS in sandy surficial aquifers, we found that DON originating from the OWTS was mobile and contributed to elevated TDN concentrations along the groundwater flowpath to the estuary. Elevated concentrations of DON in groundwater were more common during wet periods. These results suggest that if future sea level rise results in shallower groundwater tables in coastal settings, there may be an increase in OWTS DON transport. Because of the linkages between groundwater recharge and OWTS N exports, future work is needed to help quantify the potential climate change impacts on OWTS treatment processes. The sandy coastal aquifers that span much of the Atlantic Coastal Plain have shallow water tables that are generally less than 2.5 m deep (Fan et al. 2013). OWTS in these settings are sensitive to changes in precipitation excess and groundwater recharge. A better understanding of the temporal variability of nutrients from onsite and centralized wastewater treatment systems is needed to help guide sustainable development in nutrient-sensitive coastal settings.

## Acknowledgments

We thank the homeowners who allowed us access to their private residence, and permitted us to install piezometers and conduct sampling visits. We also thank the National Environmental Health Association (NEHA) for funding the study through the CDC-NEHA Contract No. 200-2008-28224. At NEHA, Tom Dickey, helped to administer the grant and was extremely supportive of our efforts. In addition, scientists from the CDC provided several thoughtful reviews of the manuscript that helped to improve it. At ECU, we are thankful for the lab analyses and support that was provided by Dr. Becky Cooper, Ms. Colleen Rochelle, and Mrs. Laura McKenna, who performed nutrient analyses. UC-Davis provided isotopic analyses. Numerous students and staff at ECU helped with our field and lab sampling efforts, including Colin Walker, Sarah Hardison, Shawn Thieme, Keaton Henry, Robert Howard, Matthew Smith, Dr. Alex Manda, Alexandria Conerly, Marcus Branch, and Alexander Jiamachello. We are especially grateful to Jim Watson and John Woods, ECU Geological Sciences, who together helped us with field logistics and sampling events. In addition, Roland Coburn provided invaluable sampling support for the first half of the study. Finally, we thank the Beaufort County Cooperative Extension and Beaufort County Health Department for their assistance in implementing this study.

## **Abbreviations**

**DON** dissolved organic nitrogen

**ET** evapotranspiration

NO<sub>3</sub><sup>-</sup> nitrate

NH<sub>4</sub><sup>+</sup> ammonium

**OWTS** onsite wastewater treatment systems

P precipitation

**TDN** total dissolved nitrogen

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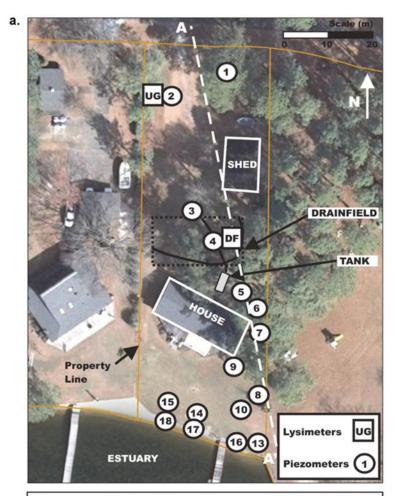
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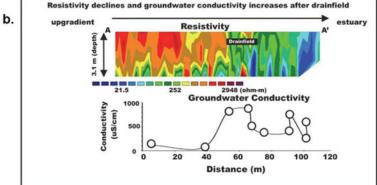
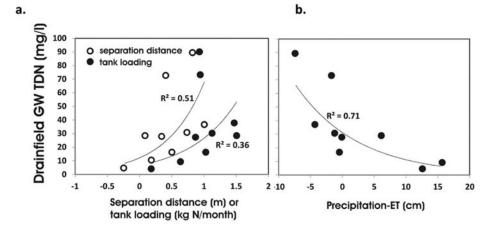
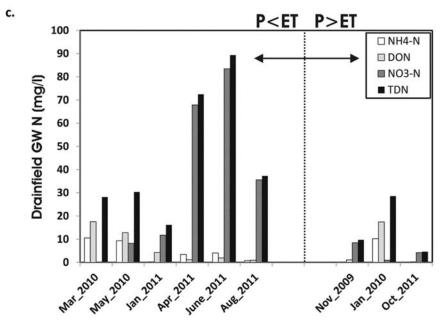


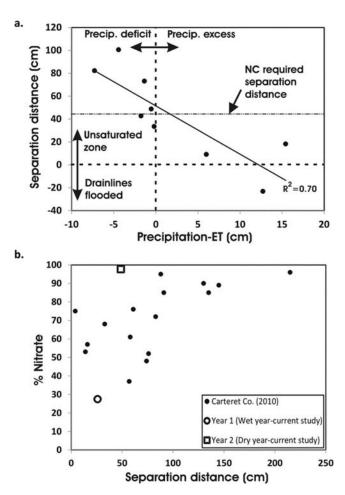
Figure 1.

**a.** Study site adjacent to the Pamlico River Estuary showing locations of piezometers (circles) and lysimeters (boxes: DF=drainfield; UG= upgradient/ background). The dashed black line indicates approximate extent of the drainfield and solid black lines indicate drainpipes. Piezometers 2, 4, 5, 6, 7, 8, 9, 10, 13, 14, and 15 were nested and contained a deep and shallow piezometer. The white dashed line from A-A' is the location of the resistivity transect. **b.** Electrical resistivity survey (Feb. 2, 2011) from A-A'. The resistivity data (ohm-m) is underlain by piezometer groundwater conductivity data.

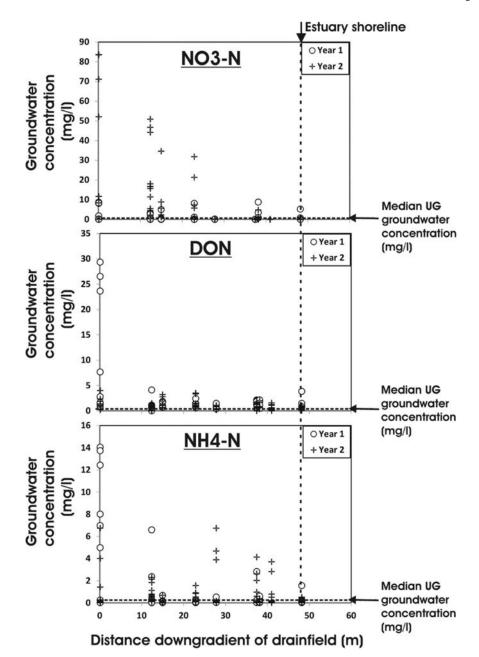




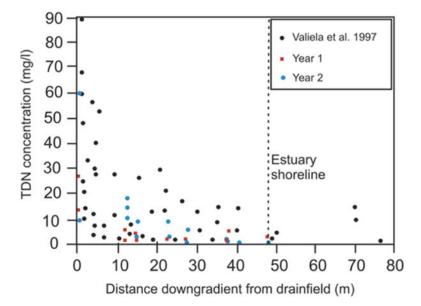
**Figure 2. a.** Separation distance (m) [ $y=12.147 e^{1.713x}$ ] or tank loading (kg-N/month) [ $y=6.722 e^{1.374x}$ ] versus the drainfield groundwater TDN concentrations. **b.** Effective precipitation (Precipitation-evapotranspiration (cm)) for two weeks prior to sampling versus the drainfield groundwater TDN concentrations [ $y=31.231 e^{-0.102x}$ ]. **c.** Drainfield groundwater nitrogen concentrations grouped by sampling dates during periods of precipitation deficits (P<ET) or precipitation excess (P>ET).



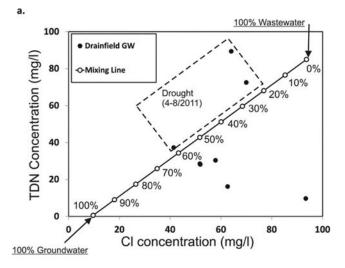
**Figure 3. a.** Relationship between effective precipitation (P-ET) for two weeks prior to sampling and separation distance. **b.** Separation distance vs. annual average % of N that occurred in drainfield groundwater as nitrate for the current study and a recent study by Humphrey et al. (2010a) in Craven County, NC.

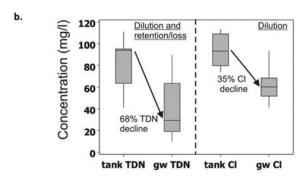


**Figure 4.**Nitrogen species concentration in groundwater related to distance from drainfield.
Upgradient (UG) concentrations were measured at upgradient piezometers (1 and 2).



**Figure 5.** Measured TDN concentrations versus distance from drainfield for Year 1 (red circles) and Year 2 (blue circles) compared to a range of values documented in an earlier study by Valiela et al. (1997).





**Figure 6. a.** Chloride and TDN concentrations in drainfield groundwater plotted with 2-component mixing model line (components of wastewater and background groundwater). **b.** The boxplots display the 25<sup>th</sup> to 75<sup>th</sup> percentiles within the box; the median is indicated by the line inside the box. The whiskers extend to the upper and lower limits of the distribution, and asterisks indicate unusually large or small outliers. The boxplots show the declines in TDN and CL between the tank (wastewater) and drainfield groundwater.

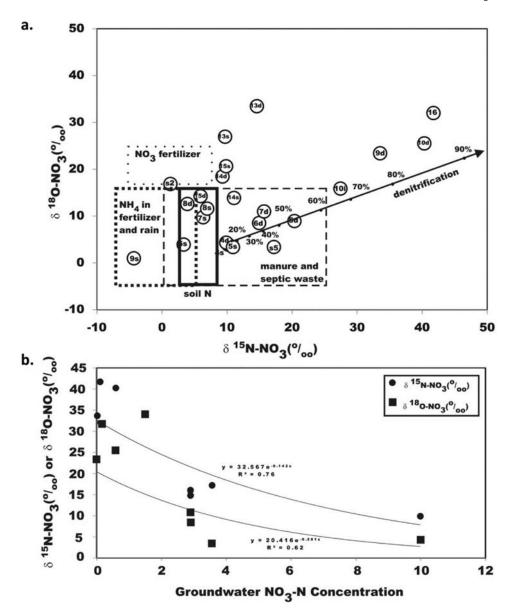
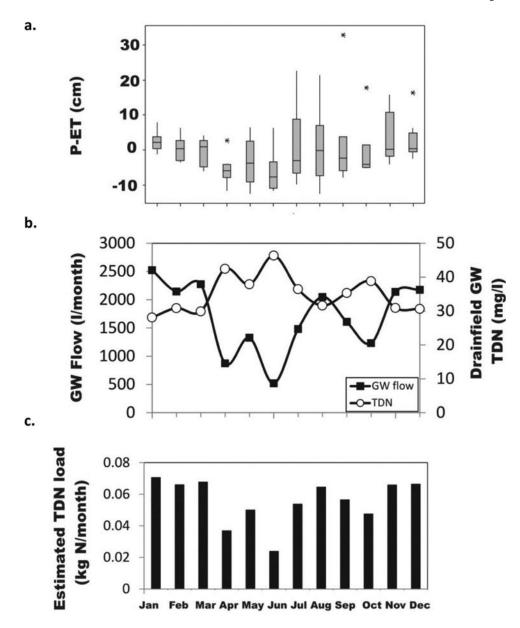


Fig. 7. (a) Enrichment in median  $^{15}$ N and  $^{18}$ O in NO $_3^-$  indicates a groundwater flow path from drainfield (1p4) to the piezometer bordering the estuary (1p16). The relationship between  $^{15}$ N and  $^{18}$ O suggests that the NO $_3^-$  source in piezometers 4d, 5d, 6d, 7d, 9d, 10d, and 16 is from the wastewater effluent and as denitrification progresses along the groundwater flowpath progressive enrichment of  $^{15}$ N and  $^{18}$ O occur in the NO $_3^-$ . (b) Median groundwater NO $_3^-$  vs.  $^{15}$ N and  $^{18}$ O patterns along a groundwater flowpath from the drainfield to the estuary (piezometers 4d, 5d, 6d, 7d, 9d, 10d, and 16) on the dates of isotopic sampling.



**Fig. 8.**(a) Monthly effective precipitation (precipitation-ET) at Plymouth, NC from 2004–2012. The boxplots display the 25<sup>th</sup> to 75<sup>th</sup> percentiles within the box; the median is indicated by the line inside the box. The whiskers extend to the upper and lower limits of the distribution, and asterisks indicate unusually large or small outliers. (b) Predicted monthly groundwater flow and drainfield groundwater TDN concentration. **c.** Predicted monthly TDN load.

#### Table 1

**a.** Summary of mean (standard deviation) of water quality parameters for upgradient groundwater (UG; piezometers 1 and 2), septic tank wastewater (ST), drainfield groundwater (DF; piezometers 4s and d), groundwater < 15 m from the drainfield (<15 m; piezometers 4, 5, and 6), and groundwater > 15 m from the drainfield (>15 m; piezometers 7,8,9,10, 13, 14, 15, 16, 17, and 18). \*due to flooding from Hurricane Irene, the October 3, 2011 soil water was affected by brackish waters from the estuary that elevated the specific conductance, if this date was excluded, UG soil water specific conductivity would be 0.034 and DF soil water 0.67 mS/cm. **b.** Summary of median N concentrations (in mg/l). Mann-Whitney test results compared the groundwater N concentrations upgradient (UG) from the drainfield with groundwater in the drainfield [DF], and downgradient from the drainfield [GW<15m and GW>15m] (italics indicates significant differences at p<0.0125; p-value for significance, adjusted for multiple comparisons using the Bonferroni method for four comparisons [p=0.05/4=0.0125] as Abdi recommended in 2007).

a.								
Source (n)	Temp(C)	pH	$D.O.\ (mg/l)$	Sp.Cond. (mS/cm)	$NH_4^+$ - $N (mg/l)$	$NO_3$ - $N (mg/l)$	DON (mg/l)	TDN  (mg/l)
<u>Wastewater</u>								
ST (13)	18.8 (4.6)	6.50 (1.0)	0.37 (0.2)	1.64 (1.5)	17.3 (5.9)	0.01 (0.0)	68.2 (18)	85.5 (18)
Soil Water								
UG (10)	16.9 (3.8)	6.8 (0.4)	7.3 (1.2)	0.85 (2.1)*	0.03 (0.0)	0.04 (0.0)	0.24 (0.1)	0.27 (0.2)
DF (12)	15.0 (4.6)	6.9 (0.5)	6.2 (1.5)	1.04 (0.8)*	0.79 (1.3)	22.3 (21)	0.92 (0.9)	28.2 (23)
<u>Groundwater</u>								
UG (15)	14.9 (4.6)	6.38 (0.7)	4.35 (1.8)	0.28 (0.6)	0.24 (0.2)	0.05 (0.1)	0.29 (0.2)	0.59 (0.4)
DF (17)	15.6 (4.3)	6.41 (0.6)	2.79 (1.4)	0.98 (0.4)	4.32 (5.2)	21.0 (31)	6.28 (9.9)	31.6 (29)
< 15 m (46)	16.5 (4.0)	6.53 (0.6)	2.35 (1.1)	0.99 (1.1)	1.93 (3.6)	13.1 (22)	2.81 (6.2)	17.6 (23)
> 15 m (101)	17.8 (5.7)	6.52 (0.6)	2.22 (1.0)	1.76 (2.8)	0.53 (1.1)	1.09 (4.1)	0.92 (0.7)	2.55 (4.4)

N-Species	UG	DF (median)	GW<15m	GW>15m	Mann-Whitney Test		
$NH_4^+$ - $N$	0.17	1.43	0.27	0.11	DF>UG (p=0.03)	<15m >UG (p=0.08)	>15m>UG (p=0.99)
$NO_3^-$ - $N$	0.03	8.28	3.86	0.05	DF>UG (p<0.01)	<15m>UG (p<0.01)	>15m>UG (p=0.14)
DON	0.28	1.63	1.00	0.77	DF>UG (p<0.01)	<15m>UG (p<0.01)	>15m>UG (p<0.01)
TDN	0.56	16.0	8.71	1.24	DF>UG (p<0.01)	<15m>UG (p<0.01)	>15m>UG (p<0.01)